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Dirac's wave equation for the electron and geometrical optics

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It will be shown how the Ansatz of WENTZEL and BRILLOUIN can be generalized in order to be applicable to the case in which the wave function possesses several components, which it does in DIRAC's theory. In that way, the approximation process will automatically imply that the diffraction effects and the effects of spin will have the same order of magnitude, as BOHR has emphasized. The geometrical-optical rays that follow from the wave equation in the limit will correspond to the classical-relativistic mechanical paths of a point electron without spin. The calculations will be carried out in the case of a one-dimensional electric field of an arbitrary character, and that will imply that the Klein paradox corresponds to a general formula for the frequency at which electrons pass into another domain in which their paths must belong to a negative mass through an intermediate region that that is classically unattainable for them.

§ 1. – Introduction.

In non-relativistic wave mechanics, the passage to the limit of classical mechanics – or, what amounts to the same thing, to geometrical optics – has already been examined in detail. With WENTZEL and BRILLOUIN, one makes the Ansatz for the solution $\psi(q, t)$ of the Schrödinger that ([1]):

$$\psi = e^{iS/h}$$

and develops S in powers of h / i:

$$S = S_0 + \frac{h}{i} S_1 + \left(\frac{h}{i}\right)^2 S_2 + \dots$$

In the zeroth approximation, one then gets the Hamilton-Jacobi partial differential equation (we abbreviate this to H.-J. eq.) from classical mechanics for S_0 , and in the first approximation, one gets a relation that is equivalent to the continuity equation by which the components of the current density and the probability density $\psi^* \psi$ are given in terms of the velocity components of the classical paths; they are equal to:

$$\dot{q}_k = \frac{\partial H}{\partial p_k},$$

([1]) Here, we refer to *h divided by* 2π as Planck's constant.

in which the expressions $p_k = \partial S_0 / \partial p_k$ have been substituted for the p_k in the Hamiltonian function $H(p, q)$. In that way, it is shown that the geometrical-optical rays that correspond to the wave-mechanical problem considered coincide precisely with the paths of the associated classical-mechanical problem.

As KRAMERS ([1]) first showed flawlessly, the method can be constructed in such a way that it also permits the asymptotic calculation of the energy eigenvalues in the limiting case of large quantum numbers. In that way, one must also employ the solutions to the H.-J. eq. in spatial regions in which one cannot arrive at the value in question of the energy E of the particle according to classical mechanics, and in which the solution S_0 of the H.-J eq. will be imaginary. The approximate solutions $\psi(q, t)$ that correspond to geometrical optics will become singular at the regression points of the classical paths where the two regions approach each other, and geometrical optics will then break down at those points. In order to find the correct connection (viz., "transition relations") between the solutions of geometrical optics in the classically-reachable region and the classical-unreachable one (that is, the particular solutions of the H.-J. eq. that approximate *the same* particular solution to the rigorous wave equation), it will not be permissible then to revert to the rigorous wave equation, as KRAMERS did. The same thing is also true in the domain of continuous eigenvalue spectra, in which one is not dealing with a determination of the eigenvalues, but the behavior of the eigenfunction in all of space.

The goal of the present communication is to adapt this method to DIRAC's relativistic wave equation for an elementary particle (e.g., electron or proton) in a given external electromagnetic potential field. That will yield some results that are characteristic of this case that do not appear in non-relativistic wave mechanics. Initially, the wave function in DIRAC's theory depends upon an index ρ whose value runs from 1 to 4, in addition to the space-time coordinates x_1, x_2, x_3, t. In order to carry out the passage to the limit of geometrical optics, one must set:

$$\psi_\rho = e^{iS_\rho / h} \tag{1}$$

here, with the special convention that *the first term* S_0 in the development:

$$S_\rho = S_0 + \frac{h}{i} S_{1\rho} + \left(\frac{h}{i}\right)^2 S_{2\rho} + \dots \tag{2}$$

does not depend upon the index ρ. Namely, it shows that one can get only solutions that correspond to the behavior of classical-mechanical paths (i.e., rays) in that way. Here, one must understand "classical" mechanics to always mean "including the relativistic corrections," since we shall introduce no restrictions about the magnitude of the speed of the electron. The theory is then applicable to – e.g. – classically-describable deflection experiments with cathode rays and β-rays in electric and magnetic fields of the type that would establish the dependency of the electron mass upon the velocity. Moreover, it is

([1]) H. A. KRAMERS, Zeit. Phys **39** (1926), 828.

formally more convenient to introduce another notation in place of (1) and (2), in such a way that one sets:

$$a_\rho = e^{S_{1\rho} + iS_{2\rho}/h + \cdots},$$

so the index 0 can then be omitted for S_0. One will then have the Ansatz:

$$\psi_\rho = a_\rho e^{iS/h}, \tag{3}$$

with

$$a_\rho = a_{0\rho} + \frac{h}{i} a_{1\rho} + \ldots, \tag{4}$$

which is completely equivalent to (1) and (2). Initially, nothing else is assumed about the reality properties of a_ρ and S in this.

The formal exposition of this Ansatz will be given in the following § 2. It is analogous to the transition from classical wave optics to geometrical optics in anisotropic media (e.g., crystals), where the polarization of light (i.e., the vector character of the field strengths) must be taken into account. The classical-mechanical paths that this passage to the limit yields in our case will be precisely the ones in relativistic mechanics for a point-electron of charge *e without the spin of the electron coming to light as a result.* That is based upon the fact that the spin moment of the electron is proportional to the quantum of action, and as a result, with our approximation, *all effects on the space-time behavior of the density and current of the particle that arise from spin must first appear automatically in the same approximation as the effects that arise from the diffraction of waves.* That is consistent with the situation that BOHR pointed out that the determination of the spin moment of a free electron is not fundamentally possible ([1]) by way of classically-describable deflection experiments. *This situation that BOHR emphasized will be automatically taken into account in the systematic approximation process that is presented here.*

Whereas the integration of the differential equations for S and $a_{0\rho}$ is not technically possible in the general case (that is, for an external electromagnetic field of an arbitrary space-time character), it can be achieved in some special cases, the most important of which is that of an electric field of well-defined direction (i.e., the one-dimensional problem in the absence of a magnetic field). That case will be developed as an example in § 3.

Furthermore, it will admit an interesting application to the KLEIN paradox, according to which, electrons can get over potential jumps with heights of order of magnitude mc^2 and continue past those potential jumps with negative mass on the basis of the theory. Here, for a given total energy E of the electron, one will be dealing with a region that is reachable for electrons of positive region according to classical relativistic mechanics and another one that is reachable for electrons of negative mass. Although those regions are separated by a classically impassable intermediate region, there will exist a finite transmission for the intermediate region in wave mechanics. In § 4, it will be shown how appealing to an imaginary solution for S in the intermediate region and the

([1]) For a discussion of several examples, as well as further references, cf., the seminar talk by W. Pauli on the magnetic electron in the Rapport du Conseil Solvay 1930.

aforementioned method of KRAMER for ascertaining the transition relations at the separation points between the regions will allow one to derive a general expression for the transmission coefficient *D*. That will demand the (asymptotic) validity of an arbitrary *continuous* behavior of the potentials and field strengths (which depend upon *only one* of the coordinates). While KLEIN's original calculation ([1]) referred to a potential *jump* at a boundary surface, which would not fall within the assumptions that are made here then, the expressions for the transmission coefficient *D* that have been obtained for special continuous potentials (e.g., for a homogeneous electric field) in the literature up to now can be obtained easily from out general expression by specialization.

§ 2. – The general approximation process.

In order to carry out our program, we shall introduce the Ansatz that we suggested before, namely:

$$\psi_\rho = a_\rho \, e^{iS/\hbar}, \tag{3}$$

$$a_\rho = a_{0\rho} + \frac{\hbar}{i} a_{1\rho} + \ldots, \tag{4}$$

into the Dirac wave equation:

$$\left(\frac{\hbar}{i} \frac{\partial}{\partial x_0} - e\,\Phi_0 \right) \psi_\rho + \alpha^k_{\rho\sigma} \left(\frac{\hbar}{i} \frac{\partial}{\partial x_k} + e\,\Phi_k \right) \psi_0 + m_0\, c\, \beta_{\rho\sigma}\, \psi_\sigma = 0. \tag{5}$$

In this, $x_0 = ct$, $\alpha^k_{\rho\sigma}$ and $\beta_{\rho\sigma}$ (k = 1 to 3; ρ, σ = 1 to 4) are the matrix elements of α^k and β, which satisfy the known relations:

$$\alpha^i \alpha^k + \alpha^k \alpha^i = 2\,\delta_{ik}\,, \qquad \alpha^k \beta + \beta \alpha^k = 0, \qquad \beta^2 = 1, \tag{6}$$

and hereinafter any doubled index will be summed over, and indeed a Latin index will be summed from 1 to 3, while a Greek one will be summed from 1 to 4; as usual, the electron charge is denoted by $-e$. Φ_0 is the electrostatic potential of our field, and Φ_k is the vector potential. By substituting (3), (4) in (5), and ordering things in powers of $\hbar/i$, while introducing the abbreviations:

$$\pi_0 = -\frac{\partial S}{\partial x_0} + \frac{e}{c}\Phi_0\,, \qquad \pi_k = \frac{\partial S}{\partial x_0} + \frac{e}{c}\Phi_k\,, \tag{7}$$

the equations will follow:

$$-\pi_0\, a_{0\rho} + \pi_k\, \alpha^k_{\rho\sigma}\, a_{0\rho} + m_0\, c\, \beta_{\rho\sigma}\, a_{0\sigma} = 0, \tag{I_0}$$

$$-\pi_0\, a_{1\rho} + \pi_k\, \alpha^k_{\rho\sigma}\, a_{1\rho} + m_0\, c\, \beta_{\rho\sigma}\, a_{1\sigma} = -\left(\frac{\partial a_{0\rho}}{\partial x_0} + \alpha^k_{\rho\sigma} \frac{\partial a_{0\sigma}}{\partial x_k} \right), \tag{I_1}$$

([1]) O. KLEIN, Zeit. Phys. **53** (1929), 157.

$$\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots,$$

$$-\pi_0\, a_{n\rho} + \pi_k\, \alpha^k_{\rho\sigma}\, a_{n\rho} + m_0\, c\, \beta_{\rho\sigma}\, a_{n\sigma} = -\left(\frac{\partial a_{n-1,\rho}}{\partial x_0} + \alpha^k_{\rho\sigma}\, \frac{\partial a_{n-1,\sigma}}{\partial x_k} \right), \qquad (\mathrm{I}_n)$$

which must be solved in succession.

If we begin with the zero-order condition (I_0) then we will see that those homogeneous equations for the $a_{0\rho}$ will possess solutions only if the π_0, π_k fulfill certain conditions that are equivalent to the vanishing of the determinant of the system of equations. In our case, one will get them in a known way by multiplying (I_0) by:

$$\pi_0\, a_{\tau\rho} + \pi_i\, \alpha^k_{\tau\sigma} + m_0\, c\, \beta_{\tau\sigma}$$

and summing over ρ, which will have:

$$(-\pi_0^2 + \sum_{k=1}^{3} \pi_k^2 + m_0^2\, c^2)\, a_{0\tau} = 0$$

as a consequence, according to the commutation relations (6). If not all $a_{0\tau}$ (i.e., for all τ) are to vanish identically then it will follow that:

$$-\pi_0^2 + \sum_{k=1}^{3} \pi_k^2 + m_0^2\, c^2 \; = 0, \qquad (\mathrm{II}_0)$$

which will coincide with the H.-J. eq. of classical-relativistic point mechanics, according to the meaning (7) of π_0 and π_k. *That therefore seems to be the solubility condition for equations* (I_0) *for the* $a_{0\rho}$.

We now imagine that S [and therefore, according to (7), π_0 and π_k] are given by any special solution of (II_0) and ask how to successively determine the $a_{0\rho}$, $a_{1\rho}$, etc. In order to do that, it is essential that the $a_{0\rho}$ should still not be determined completely by (I_0). It is known that for given π_0 and π_k, there exist two linearly-independent solutions of (I_0):

$$a_{0\rho} = A_\rho\,(\pi_0\,,\, \pi_k) \qquad \text{and} \qquad a_{0\rho} = B_\rho\,(\pi_0\,,\, \pi_k)$$

that correspond to the two spin directions. One can think of them as having been chosen (cf., also § 3 on this subject) such that they do not depend upon the coordinates x_0, x_k explicitly, but only on the π_0, π_k themselves (which are functions of the coordinates in their own right), which we shall express by the notation. However, the general solution of (I_0) will then be:

$$a_{0\rho} = C\,(x_0, x_k)\, A_\rho\,(\pi_0\,,\, \pi_k) + C'(x_0, x_k)\, B_\rho\,(\pi_0\,,\, \pi_k), \qquad (8)$$

in which C and C' are initially two arbitrary functions of the coordinates (that are independent of the index ρ).

However, one should observe that *the condition for the solubility of equations* (I_1) *in the following approximation* will impose further restrictions on the $a_{0\rho}$. Here, one is

indeed dealing with an inhomogeneous linear system of equations whose associated homogeneous equations possess solutions. In this case, the solubility condition for the inhomogeneous system is known to be that its right-hand side must be orthogonal to the adjoint homogeneous equations. In our case (for σ = 1 to 4), the latter reads:

$$-a_\rho^+ \pi_0 + a_\rho^+ \alpha_{\rho\sigma}^k \pi_k + a_\rho^+ \beta_{\rho\sigma} m_0 c = 0. \tag{9}$$

If a system of numbers a_ρ^+ satisfies these equations then, in fact, one will have:

$$\sum_\rho a_\rho^+ (-\pi_0 a_{n\rho} + \pi_k \alpha_{\rho\sigma}^k a_{n\sigma} + m_0 c \beta_{\rho\sigma} a_{n\sigma}) \equiv 0$$

identically in the $a_{\nu\rho}$, so the further condition that:

$$a_\rho^+ \left(\frac{\partial a_{0\rho}}{\partial x_0} + \alpha_{\rho\sigma}^k \frac{\partial a_{0\sigma}}{\partial x_k} \right) = 0, \tag{II$_1$}$$

$$\ldots\ldots\ldots\ldots\ldots\ldots\ldots,$$

$$a_\rho^+ \left(\frac{\partial a_{n-1,\rho}}{\partial x_0} + \alpha_{\rho\sigma}^k \frac{\partial a_{n-1,\sigma}}{\partial x_k} \right) = 0 \tag{II$_1$}$$

must follow from (I$_1$), …, (I$_n$).

Since equations (9), like (I$_0$), possess two linearly-independent solutions (which we will give shortly), (II$_1$) will split into two independent equations *that can be regarded as two simultaneous first-order partial differential equations for the still-undetermined remaining functions C and C´ in* (8). If the C and C' are given arbitrarily at a well-defined time point then equations (II$_1$) will determine their further time evolution uniquely. In general, the solubility conditions (II$_n$) for equations (I$_n$) serve to determine the $a_{n-1,\rho}$ completely.

We can give the solutions of (9) immediately, since the α^k and β are Hermitian matrices. If $A_\rho(\pi_0, \pi_k)$ and $B_\rho(\pi_0, \pi_k)$ are the solutions of (I$_0$) that were introduced before – and indeed identically in π_0 and π_k, insofar as only (II$_0$) is fulfilled – then we can fulfill relations (9) with:

$$a_{0\rho}^+ = A_\rho^*(\pi_0, \pi_k) \qquad \text{and} \qquad a_{0\rho}^+ = B_\rho^*(\pi_0, \pi_k).$$

Therefore, A_ρ^* and B_ρ^* are complex conjugates of the functions A_ρ and B_ρ ; that should say that anywhere i occurs explicitly, it will be replaced with – i. However, for the case in which π_0 and π_k are not real, they are *not* replaced with their complex-conjugate values, but will keep their original values. It is only for real π_0 and π_k that A_ρ^* and B_ρ^* will have numerical values that are complex-conjugate to A_ρ and B_ρ. In place of (II$_1$), …, (II$_n$), we can also set:

$$\left.\begin{aligned} A^*_\rho(\pi_0,\pi_k)\left(\frac{\partial a_{0\rho}}{\partial x_0}+\alpha^k_{\rho\sigma}\frac{\partial a_{0\sigma}}{\partial x_k}\right)=0,\\ B^*_\rho(\pi_0,\pi_k)\left(\frac{\partial a_{0\rho}}{\partial x_0}+\alpha^k_{\rho\sigma}\frac{\partial a_{0\sigma}}{\partial x_k}\right)=0,\end{aligned}\right\} \qquad (\mathrm{II}'_1)$$

$$\text{……..,}$$

$$\left.\begin{aligned} A^*_\rho(\pi_0,\pi_k)\left(\frac{\partial a_{n-1,\rho}}{\partial x_0}+\alpha^k_{\rho\sigma}\frac{\partial a_{n-1,\sigma}}{\partial x_k}\right)=0,\\ B^*_\rho(\pi_0,\pi_k)\left(\frac{\partial a_{n-1,\rho}}{\partial x_0}+\alpha^k_{\rho\sigma}\frac{\partial a_{n-1,\sigma}}{\partial x_k}\right)=0.\end{aligned}\right\} \qquad (\mathrm{II}'_n)$$

One will also get relations that are equivalent to these when one multiplies the relations (I_1), …, (I_n) by:

$$+\pi_0\,\delta_{\tau\rho}+\pi_l\,\alpha^l_{\tau\rho}+m_0\,c\,\beta_{\tau\rho}$$

and sums over ρ, in complete analogy to the transition from (I_0) to (II_0). Therefore, as a result of (II_0), the left-hand side will vanish identically, and one will get:

$$+\pi_0\left(\frac{\partial a_{0\tau}}{\partial x_0}+\alpha^k_{\tau\sigma}\frac{\partial a_{0\sigma}}{\partial x_k}\right)+\pi_l\left(\alpha^l_{\tau\sigma}\frac{\partial a_{0\rho}}{\partial x_0}+(\alpha^l\alpha^k)_{\tau\sigma}\frac{\partial a_{0\sigma}}{\partial x_k}\right)+m_0c\left(\beta_{\tau\rho}\frac{\partial a_{0\rho}}{\partial x_k}+(\beta\alpha^k)_{\tau\sigma}\frac{\partial a_{0\sigma}}{\partial x_k}\right)=0, \quad (\mathrm{II}''_1)$$

$$\text{……..,}$$

$$+\pi_0\left(\frac{\partial a_{n-1,\tau}}{\partial x_0}+\alpha^k_{\tau\sigma}\frac{\partial a_{n-1,\sigma}}{\partial x_k}\right)+\pi_l\left(\alpha^l_{\tau\sigma}\frac{\partial a_{n-1,\rho}}{\partial x_0}+(\alpha^l\alpha^k)_{\tau\sigma}\frac{\partial a_{n-1,\sigma}}{\partial x_k}\right)+m_0c\left(\beta_{\tau\rho}\frac{\partial a_{n-1,\rho}}{\partial x_k}+(\beta\alpha^k)_{\tau\sigma}\frac{\partial a_{n-1,\sigma}}{\partial x_k}\right)=0. \quad (\mathrm{II}''_n)$$

In that way, we indeed get four equations (II''_n) for each n, but only two of them will be linearly-independent, and they will be equivalent to (II'_n).

We have not succeeded in integrating equations (I) and the conditions (II) that follow from them in the general case (that is, for an arbitrary external field Φ_k). Rather, an important consequence of equations (I''_1) and (II''_1) should be discussed that exhibits the relationship between those equations and the classical-mechanical paths. S (and therefore π_0 , π_k , as well) will be real in the classically-reachable region, and that might be assumed explicitly for the following considerations (in contrast to the ones up to now). When one employs the assumed reality of π_0 , π_k , it will next follow from (II'_n) and (8) that:

$$a^*_{0\rho}\left(\frac{\partial a_{0\rho}}{\partial x_0}+\alpha^k_{\rho\sigma}\frac{\partial a_{0\sigma}}{\partial x_k}\right)=0,$$

and due to the Hermiticity of the α^k, when one goes to the complex conjugate, one will get:

$$\left(\frac{\partial a_{0\sigma}^{*}}{\partial x_0} + \frac{\partial a_{0\rho}^{*}}{\partial x_k} \alpha_{\rho\sigma}^{k} \right) a_{0\rho} = 0.$$

Adding these two equations and introducing density and current according to:

$$\rho = a_{0\rho}^{*} a_{0\rho}, \quad s_k = \frac{1}{c} i_k = a_{0\rho}^{*} \alpha_{\rho\sigma}^{k} a_{0\sigma} \tag{10}$$

will imply the continuity equation:

$$\frac{\partial \rho}{\partial x_0} + \frac{\partial s_k}{\partial x_k} = 0. \tag{11}$$

One observes that according to (3) for real S, this function will drop out of the expressions for density and current, such that the latter depends upon only a_ρ ; the expressions (10) are then those of the first approximation. Furthermore, it should be stressed that the continuity equation says less than equations (I_1), (II_1'), such that the latter cannot be deduced from the former.

We can now derive a simple expression for the quotients of the current and density that coincides in an entirely analogous way with the one in non-relativistic wave mechanics for the velocity of the electron on a classical path in the approximation considered. We then start from the relations (I_0) and (9) and replace a_ρ^{+} with $a_{0\rho}^{*}$ in the latter, which is permissible for real π_0, π_k. We write those relations in the form:

$$\begin{aligned} &- \pi_0 a_{0\sigma} + \pi_l \alpha_{\sigma\tau}^{l} a_{0\tau} + m_0 c \beta_{\sigma\tau} a_{0\tau} = 0, \\ &- a_{0\rho}^{*} \pi_0 + a_{0\rho}^{*} \alpha_{\rho\sigma}^{l} \pi_l + a_{0\sigma}^{*} \beta_{\rho\sigma} m_0 c = 0, \end{aligned}$$

multiply the first equation on the left by $a_{0\rho}^{*} \alpha_{\rho\sigma}^{k}$, the second equation on the right by $\alpha_{\sigma\tau}^{k} a_{0\tau}$, sum over σ both times, and add both equations. We will then get:

$$- 2\pi_0 (a_{0\rho}^{*} \alpha_{\rho\sigma}^{k} a_{0\sigma}) + \pi_l a_{0\rho}^{*} (\alpha^k \alpha^l + \alpha^l \alpha^k)_{\rho\tau} a_{0\tau} + m_0 c\, a_{0\rho}^{*} (\alpha^k \beta + \beta \alpha^k)_{\rho\tau} a_{0\tau} = 0.$$

With consideration given to the commutation relations (6) and the use of the expressions (10) for density and current, this will simplify to:

$$\pi_0 s_k = \pi_k \rho$$

or

$$i_k = c\, s_k = \rho \frac{c \pi_k}{\pi_\rho}. \tag{12}$$

The Hamilton function $H(p_k, x_k)$ of classical-relativistic point-mechanics is now:

$$H\,(p_k\,,\,x_k) = -\,\frac{\partial S}{\partial t}\;=\;c\left(\,\pi_0 - \frac{e}{c}\Phi_0\,\right) = \;c\sqrt{m^2c^2 + \sum_{k=1}^{3}\left(\,p_k + \frac{e}{c}\Phi_k\,\right)^2}\;\;-\,e\,\Phi_0\,, \tag{13}$$

so:

$$\dot{x}_k \;=\; \frac{\partial H}{\partial p_k} \;=\; \frac{c\,\pi_k}{\sqrt{m^2c^2 + \sum_{k=1}^{3}\pi_k^2}} \;=\; \frac{c\,\pi_k}{\pi_0}\,. \tag{14}$$

In this, in accord with (7) and (II_0), one has set:

$$\pi_0 = +\sqrt{m^2c^2 + \sum_{k=1}^{3}\pi_k^2}\;. \tag{15}$$

As one sees, this choice of sign in the square root in (13), (14), (15) corresponds to a particle of positive mass. For a particle of negative mass, one would have:

$$H\,(p_k\,,\,x_k) = -\,\frac{\partial S}{\partial t}\;=\;c\left(\,\pi_0 - \frac{e}{c}\Phi_0\,\right) = -\;c\sqrt{m^2c^2 + \sum_{k=1}^{3}\left(\,p_k + \frac{e}{c}\Phi_k\,\right)^2}\;\;-\,e\,\Phi_0\,, \tag{13′}$$

$$\dot{x}_k \;=\; \frac{\partial H}{\partial p_k} \;=\; -\frac{c\,\pi_k}{\sqrt{m^2c^2 + \sum_{k=1}^{3}\pi_k^2}} \;=\; \frac{c\,\pi_k}{\pi_0}\,, \tag{14′}$$

$$\pi_0 = -\sqrt{m^2c^2 + \sum_{k=1}^{3}\pi_k^2}\;. \tag{15′}$$

A comparison of (14) and (14′) with (12) will show that in both cases, one has:

$$i_k = \rho\,\frac{\partial H}{\partial p_k} \;=\; \rho\,\dot{x}_k\;.$$

It will then follow from the continuity equation (11) that the particle will propagate along the paths that are determined by:

$$\dot{x}_k = \frac{\partial H}{\partial p_k}\,. \tag{16a}$$

From the H.-J. eq. (II_0), which is equivalent to:

$$-\,\frac{\partial S}{\partial t} = H\left(\frac{\partial S}{\partial x_k}, x_k\right),$$

and from:

$$p_k = \frac{\partial S}{\partial x_k},$$

it will then follow in a known way that:

$$\dot{p}_k = \frac{\partial^2 S}{\partial x_k \partial t} + \frac{\partial^2 S}{\partial x_k \partial x_l}\dot{x}_l = -\left(\frac{\partial H}{\partial x_k}\right)_p - \frac{\partial H}{\partial p_l}\frac{\partial^2 S}{\partial x_l \partial x_k} + \frac{\partial^2 S}{\partial x_k \partial x_l}\frac{\partial H}{\partial p_l},$$

so

$$\dot{p}_k = -\left(\frac{\partial H}{\partial x_k}\right)_p. \tag{16b}$$

The paths are then precisely those of classical-relativistic point mechanics, and indeed for a particle without spin. The effects that originate in spin (at least, as far as their influence on the evolution of density and current is concerned) are first contained in the amplitudes $a_{1\rho}$ in the next approximation along with the diffraction effects, but we would not like to go further into their computation.

A special case of the general solution of (I) and (II) is that of the stationary solution, in which time is contained only as an exponential function according to:

$$\psi_\rho = u_\rho(s)\ e^{-iEt/\hbar}, \tag{17}$$

in which E then means the energy. One must then set:

$$S = \bar{S}(x) - Et \tag{18}$$

and

$$a_\rho = a_\rho(x), \qquad \frac{\partial a_\rho}{\partial x_0} = 0,$$

$$u_\rho(x) = a_\rho(x)\ e^{-i\bar{S}/\hbar}, \tag{3′}$$

in which $\bar{S}$ and a will now be independent of t. In place of (7), one finds:

$$\pi_0 = \frac{E + e\Phi_0}{c}, \qquad \pi_k = \frac{\partial \bar{S}}{\partial x_k}, \tag{7′}$$

$$H\left(\frac{\partial \bar{S}}{\partial x_k}, x_k\right) = E,$$

and all terms in equations (I) and (II) that contain differentiations with respect to x_0 will drop out.

§ 3. – Example of a particle in an electric field with a fixed direction.

We lay the x_3-axis in the direction of the field such that Φ_0 depends upon only x_3, but is otherwise arbitrary. As a result of the absence of a magnetic field, one will have $\Phi_k = 0$, so $\pi_k = p_k = \partial S / \partial x_k$. We would like to restrict ourselves to the case in which the particles move parallel to the x_3-axis, so one will have $p_1 = p_2 = 0$. For the stationary solution that is associated with the energy E, S will also depend upon only x_3, and indeed one will have:

$$\pi_0 = \frac{E + e\Phi_0}{c}, \qquad p_3 = \pi_3 = \pm\sqrt{\pi_0^2 - m^2c^2}, \qquad \overline{S} = \int^{x_3} \pi_3 \, dx_3 . \tag{19}$$

In the classically-reachable region, one will have $| \pi_0 | > mc$, so π_3 will be real, and the two signs will correspond to the two directions of motion of the particle along $+ x_3$ or $- x_3$.

For the further calculations, we now specialize the Dirac matrices; e.g., such that β will be diagonal:

$$\beta = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}, \tag{20a}$$

$$\alpha_1 = \begin{pmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix}, \quad \alpha_2 = \begin{pmatrix} 0 & 0 & 0 & -i \\ 0 & 0 & i & 0 \\ 0 & -i & 0 & 0 \\ i & 0 & 0 & 0 \end{pmatrix}, \qquad \alpha_3 = \begin{pmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \\ 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \end{pmatrix}. \tag{20b}$$

Equations (I_1) will then read:

$$\left.\begin{aligned} (-\pi_0 + mc)\, a_{01} + \pi_3 \, a_{03} = 0, \\ (-\pi_0 - mc)\, a_{03} + \pi_3 \, a_{01} = 0, \end{aligned}\right\} \tag{21a}$$

$$\left.\begin{aligned} (-\pi_0 + mc)\, a_{02} - \pi_3 \, a_{04} = 0, \\ (-\pi_0 - mc)\, a_{04} - \pi_3 \, a_{02} = 0. \end{aligned}\right\} \tag{21b}$$

By means of the continuity equation, the current component s_3 must be constant for stationary solutions in our one-dimensional problem, for which one will have:

$$\frac{\partial \rho}{\partial x_0} = 0,$$

and it is given by:

$$s_3 = (a_{01}^* a_{03} + a_{03}^* a_{01}) - (a_{02}^* a_{04} + a_{04}^* a_{02}) . \tag{22}$$

Equations (21a) and (21b) will be satisfied by the Ansatz:

$$a_{0\rho} = A_\rho \qquad \text{or} \qquad a_{0\rho} = B_\rho \tag{23}$$

when we normalize according to:

$$A_1 A_3 = 1, \qquad B_2 B_4 = -1 \tag{24}$$

and set:

$$A_1 = \sqrt{\frac{\pi_3}{\pi_0 - mc}}, \qquad A_3 = \sqrt{\frac{\pi_0 - mc}{\pi_3}}, \qquad A_2 = A_4 = 0, \tag{25a}$$

$$B_1 = B_3 = 0, \quad B_2 = -\sqrt{\frac{\pi_3}{\pi_0 - mc}}, \qquad B_4 = \sqrt{\frac{\pi_0 - mc}{\pi_3}} . \tag{25b}$$

In this, one must observe that due to $\pi_3^2 = \pi_0^2 - m^2 c^2$, one of the two equations (21a) or (21b) will already follow from the other one, and *vice versa*. From (22), the current s_3 will be constant for any of the solutions A_ρ and B_ρ for real π_0 and π_3.

We now still have to test conditions (II_1'), which read:

$$\begin{aligned} A_1 \frac{\partial a_{03}}{\partial x_3} + A_3 \frac{\partial a_{01}}{\partial x_3} &= 0, \\ -B_2 \frac{\partial a_{04}}{\partial x_3} - B_4 \frac{\partial a_{02}}{\partial x_3} &= 0, \end{aligned} \tag{26}$$

or

$$\begin{aligned} \frac{1}{A_1}\frac{\partial a_{01}}{\partial x_3} &= -\frac{1}{A_3}\frac{\partial a_{03}}{\partial x_3}, \\ \frac{1}{B_2}\frac{\partial a_{02}}{\partial x_3} &= -\frac{1}{B_4}\frac{\partial a_{04}}{\partial x_3}, \end{aligned} \tag{26}$$

in our case. (i does not enter into A_ρ and B_ρ explicitly!). These conditions are fulfilled for $a_{0\rho} = A_\rho$ and $a_{0\rho} = B_\rho$ by means of (24), such that the most general admissible solution will read:

$$a_{0\rho} = C_1 A_\rho + C_2 B_\rho \tag{27}$$

with *constant* (i.e., independent of the coordinates) C_1 and C_2 . That Ansatz is also applicable in the event that the expressions under the roots in (25a) and (25b) are not positive and real. The $a_{0\rho}$ will become singular at the places where $\pi_3 = 0$ (so $\pi_0 = \pm mc$), and the geometrical optics approximation will break down there.

§ 4. – Application to the Klein paradox.

It was already mentioned in § 2 that the case of $\pi_0 > 0$ corresponds to a particle with positive mass, while the case of $\pi_0 < 0$ corresponds to that of a particle of negative mass. If K_l means the components of the Lorentz force that acts upon the electron charge $(-e)$ for a given external electromagnetic field then it will, in fact, follow from the canonical equations of motion (16) and the Hamilton function (13) [(13′), resp.] that:

$$\frac{d}{dt}\left(\frac{m\dot{x}_l}{\sqrt{1-\sum_k \frac{\dot{x}_k^2}{c^2}}}\right) = K_l$$

or

$$-\frac{d}{dt}\left(\frac{m\dot{x}_l}{\sqrt{1-\sum_k \frac{\dot{x}_k^2}{c^2}}}\right) = K_l\,,$$

resp.

For $\pi_0 < 0$, the acceleration will then be in the opposite direction to the force, as would be the case for negative kinetic energy.

As would follow from (15) [(15′), resp.], for real π_k, one will constantly have $|\pi_0| > mc$, such that the solutions with $\pi_0 > 0$ can never go continuously over to the ones with $\pi_0 < 0$. In classical-relativistic point mechanics, the exclusion of the latter solutions, which contradict experiment, can be done in a manner that is unique and free of contradictions. However, as KLEIN is known to have shown, that is not the case in wave mechanics, since in that field, the regions in which the π_k are partially complex and π_0 lies between $-mc$ and $+mc$ can likewise be attained by particles.

We shall explain this in the example of a one-dimensional electric field that was considered in the previous section. In it, we shall imagine that the potential Φ_0 *decreases continuously* for increasing x_3, such that it will become negative for a sufficiently large x_3. For a given energy E, we will then have three regions to distinguish:

1) $x_3 < a$, $\quad mc < \pi_0 = \dfrac{E + e\Phi_0}{c}$,

2) $a < x_3 < b$, $\quad -mc < \pi_0 = \dfrac{E + e\Phi_0}{c} < mc$,

3) $b < x_3$, $\quad \pi_0 = \dfrac{E + e\Phi_0}{c} < -mc$.

$x_3 = a$ corresponds to the regression point of the classical path in region 1) of a particle with positive mass and total energy E, while the point $x_3 = b$, which lies further to the right, corresponds to the classical path in region 3) of a particle with negative mass and the same energy E. One has that $\pi_3^2 = \pi_0^2 - m^2 c^2$ is positive in region 1), as well as in region 3), so π_3 will be real. π_3^2 will be negative in the classical-unreachable region 2), so π_3 will be pure imaginary.

Now, a wave that moves to the right, as well as one that moves to the left, can exist in each region, and for that reason we must characterize the special solution of the wave equation that we would like to consider in more detail. It shall be that stationary solution that can be regarded as the limiting case of a wave packet that moves to the left from the critical point $x_3 = a$. It is characterized by the fact that *only one* wave that proceeds to the right is present in the region 3), by which we mean that the group velocity of the wave that is defined by (14′) should point towards $+ x_3$. An incident wave and a reflected one will then be present in region 1). One must then ascertain the transmission coefficient D, which is defined to be the quotient of the current of the wave that advances to the right in 3) by the current of the wave that advances to the right in 1). We will see that our entire asymptotic approximation is true only for the case in which D is a very small number. A formal simplification will be achieved in that way such that one can restrict oneself to the case in which a_{03} and a_{04} are zero throughout; i.e., to the solution A_ρ in § 3. The solution B_ρ, for which a_0 and a_{02} will vanish, as well as the most general linear combination of both solutions according to (27), would, in fact, yield exactly the same transmission coefficient, as one can easily verify.

According to (25a), corresponding to $\pi_3 = \pm\sqrt{\pi_0^2 - m^2c^2}$, we then set:

In region 1):

$$u_{01} = a_{01}\, e^{i\bar{S}/h}$$

$$= \left(\frac{\pi_0 + mc}{\pi_0 - mc}\right)^{1/4}\left[C_1 \exp\left(\frac{i}{h}\int_a^{x_3}\sqrt{\pi_0^2 - m^2c^2}\,dx_3\right) + C_1' \exp\left(-\frac{i}{h}\int_a^{x_3}\sqrt{\pi_0^2 - m^2c^2}\,dx_3\right)\right],$$

$$u_{03} = a_{03}\, e^{i\bar{S}/h}$$

$$= \left(\frac{\pi_0 - mc}{\pi_0 + mc}\right)^{1/4}\left[C_1 \exp\left(\frac{i}{h}\int_a^{x_3}\sqrt{\pi_0^2 - m^2c^2}\,dx_3\right) - C_1' \exp\left(-\frac{i}{h}\int_a^{x_3}\sqrt{\pi_0^2 - m^2c^2}\,dx_3\right)\right],$$

$$s_3 = u_{01}^* u_{03} + u_{03}^* u_{01} = 2\,(|\,C_1\,|^2 - |\,C_1'\,|^2). \qquad (28_1)$$

In region 2):

$$u_{01} = a_{01}\, e^{i\bar{S}/h}$$

$$= e^{-i\pi/4}\left(\frac{mc+\pi_0}{mc-\pi_0}\right)^{1/4}\left[C_2\exp\left(-\frac{1}{h}\int_a^{x_3}\sqrt{\pi_0^2-m^2c^2}\,dx_3\right)+C_2'\exp\left(+\frac{1}{h}\int_a^{x_3}\sqrt{\pi_0^2-m^2c^2}\,dx_3\right)\right],$$

$u_{03} = a_{03}\ e^{i\bar{S}/h}$

$$= e^{+i\pi/4}\left(\frac{mc-\pi_0}{mc+\pi_0}\right)^{1/4}\left[C_2\exp\left(-\frac{1}{h}\int_a^{x_3}\sqrt{\pi_0^2-m^2c^2}\,dx_3\right)-C_2'\exp\left(+\frac{1}{h}\int_a^{x_3}\sqrt{\pi_0^2-m^2c^2}\,dx_3\right)\right],$$

$$s_3 = u_{01}^* u_{03} + u_{03}^* u_{01} = 2i\ [C_2\, C_2^* - C_2^*\, C_2'].\qquad (28_2)$$

In region 3):

$u_{01} = a_{01}\ e^{i\bar{S}/h}$

$$= \left(\frac{-\pi_0-mc}{-\pi_0+mc}\right)^{1/4}\left[C_3\exp\left(-\frac{i}{h}\int_a^{x_3}\sqrt{\pi_0^2-m^2c^2}\,dx_3\right)+C_3'\exp\left(+\frac{i}{h}\int_a^{x_3}\sqrt{\pi_0^2-m^2c^2}\,dx_3\right)\right],$$

$u_{03} = a_{03}\ e^{i\bar{S}/h}$

$$= \left(\frac{-\pi_0+mc}{-\pi_0-mc}\right)^{1/4}\left[C_3\exp\left(-\frac{i}{h}\int_a^{x_3}\sqrt{\pi_0^2-m^2c^2}\,dx_3\right)+C_3'\exp\left(-\frac{i}{h}\int_a^{x_3}\sqrt{\pi_0^2-m^2c^2}\,dx_3\right)\right],$$

$$s_3 = u_{01}^* u_{03} + u_{03}^* u_{01} = 2\ (|\, C_3\,|^2 - |\, C_3'\,|^2).\qquad (28_3)$$

As would emerge from the expression for the current, π_3 will be negative (positive, resp.) in region 3) for the wave that advances to the right (left, resp.), in contrast to s_3 . Corresponding to our program, we will have to set $C_3' = 0$ for the solution to be considered in it.

Should our solutions approximate one and the same particular solution of the rigorous wave equation, then since the current s_3 will be consistently constant for them, s_3 must have coincident values in the three regions.

$$|\, C_1\,|^2 - |\, C_1'\,|^2 = i\ [C_2\, C_2'^* - C_2^*\, C_2'] = |\, C_3\,|^2 - |\, C_3'\,|^2 .\qquad (29)$$

The transmission coefficient is:

$$D = \frac{|\, C_3\,|^2}{|\, C_1\,|^2}\ .\qquad (30)$$

When that is a small number, $|\, C_1'\,|$ will differ only slightly from $|\, C_1\,|$, and furthermore C_2' (compared with $|\, C_1\,|^2$) will have order D, and C_2 will have order 1, since the two

terms of u_{01} and u_{03} must have the same order of magnitude in region 2) in the vicinity of $x_3 = b$.

Geometrical optics will not suffice to get the transition relations at the critical locations, and will be necessary to appeal to the rigorous wave equations, moreover. On the other hand, it must be employed only in the vicinity of critical points, but not in the intermediate region. How things will happen in that region is not characteristic of the relativistic theory, but is entirely analogous to the non-relativistic theory. For that reason, we would like to address that only briefly here and refer to the investigations of KRAMERS that were cited in the Introduction for the details. In the neighborhood of the critical location, one can either replace $\pi_0 + mc$ ($\pi_0 - mc$, resp.) with a linear process and deduce the *general* form of the transition relations from the behavior of the exact solutions of the wave equations for a *homogeneous* field at the critical points, or one can avoid the critical locations in the complex plane and examine the behavior of the asymptotic particular solutions in the complex plane. In that, one will assume of the exact solution of the wave equation only that it can be continued analytically into the complex plane in the neighborhood of critical locations. That will always be applicable when the potential and its derivative are continuous there.

If we initially consider the first critical location $x_3 = a$ then we can show by such an argument that ([1]):

$$\text{One has } C_2' = C_1' \text{ for } C_1 = 0.$$
$$\text{One has } C_2' = 0 \quad \text{for } C_2 = C_1 .$$

Moreover, since u_{01} is real and u_{03} is pure imaginary in region 1), as one would gather from the rigorous wave equation, the same thing will be true for all regions, and the linear connection between $C_1,\ C_1'$ and $C_2,\ C_2'$ will already be determined by that. One will then have:

$$\begin{aligned} C_2 &= \frac{1}{2} C_1 + \frac{i}{2} C_1', \\ C_2' &= i C_1 + C_1' . \end{aligned} \tag{31a}$$

With that, one confirms the first part of equation (29), moreover.

The treatment of the second critical location $x_3 = b$ takes an analogous form. One first introduces the following abbreviation: The action integral:

$$W = \frac{1}{h}\int_a^b |\pi_3|\, dx_3 = \frac{1}{h}\int_a^b \sqrt{m^2c^2 - \pi_0^2}\, dx_3 = \frac{1}{h}\int_a^b \sqrt{m^2c^2 - \left[\frac{E + e\Phi_0(x_3)}{c}\right]^2}\, dx_3 \tag{32}$$

between the critical locations, which will play an essential role in what follows. Hence:

$$\bar{C}_2 = C_2\, e^{-W}, \quad \bar{C}_2' = C_2'\, e^{+W}. \tag{33}$$

([1]) Cf., in particular, A. ZWAAN, Diss. Utrecht 1929, Chap. III, § 2, along with H. A. KRAMERS and G. P. ITTMANN, Zeit. Phys. **58** (1929), 217, especially pps. 221 and 222.

The latter expressions are chosen such that (28_2) will remain true when one replaces the lower integration limit a with b and simultaneously replaces C_2, C_2' with $\bar{C}_2$, $\bar{C}_2'$. One will then first get:

$$\bar{C}_2 = C_3 \text{ is true for } C_3' = 0,$$
$$\bar{C}_2 = C_3' \text{ is true for } \bar{C}_2 = 0,$$

and then, if one recalls the reality relations:

$$\bar{C}_2 = C_3 + iC_3', \qquad \bar{C}_2' = \frac{i}{2}C_3 + \frac{1}{2}C_3'. \tag{31b}$$

Generally, the second of equations (29) will be true as a consequence of (31b).

If we set $C_3' = 0$, moreover, then it follow from (31a), (33), (31b) that:

$$\frac{1}{2}C_1 + \frac{i}{2}C_1' = e^W C_3, \qquad i\,C_3 + C_1' = e^{-W} C_3, \tag{34}$$

and from this:

$$C_3 = \frac{e^{-W}}{1+\frac{1}{4}e^{-2W}} C_1, \qquad C_1' = -\,i\,\frac{1-\frac{1}{4}e^{-2W}}{1+\frac{1}{4}e^{-2W}} C_1, \tag{35}$$

from which one can easily verify the relation:

$$|C_3|^2 = |C_1|^2 - |C_1'|^2.$$

In evaluating the precision that the validity of this result can claim, one must observe that there must be a region between the points $x_3 = a$ and $x_3 = b$ whose extent is comparable to $b - a$, and in which the gradient of the wave length $2\pi h / |\pi_3|$ is small; the application of geometrical optics would not be allowed in the other case. If that is fulfilled then W will constantly be a large number, and we can and should therefore neglect e^{-2W} in comparison to 1. In place of (35), we will get simply:

$$C_3 = e^{-W} C_1, \qquad C_1' = -\,i\,C_1, \tag{35a}$$

and for the transmission coefficient that is defined by (30):

$$D = e^{-2W}. \tag{36}$$

The independence of this value of D from the polarization of the electron wave was mentioned already.

Up to now, that result has not been presented in the literature in the generality that was attained here – viz., the behavior of the potential $\Phi_0\,(x_3)$ between the critical locations was arbitrary, except for continuity. D was calculated by SAUTER ([1]) for two special assumptions about the behavior of the potential, namely, the homogeneous field:

$$-e\,\Phi_0\,(x_3) = a\,x_3$$

and the field that behaves like:

$$-e\,\Phi_0\,(x_3) = \frac{P}{2}\frac{1-e^{-a\,x_3}}{1+e^{-a\,x_3}}.$$

The evaluation of the action integral (32) in these special cases will yield exact agreement between our general expression (36) and the values of D that SAUTER gave. On that subject, it should be remarked that SAUTER also introduced asymptotic approximations for the evaluation of the rigorous solution that correspond to ours precisely.

§ 5. – Remarks on the question of the fundamental meaning of the transition to states with negative kinetic energy.

Allow us to add some remarks about the fundamental meaning of the Klein paradox here, although they will have only a tenuous connection with the foregoing methodical examination, which took place completely within the scope of the theory up to now. The author is of the opinion that all of the attempts up to now to deal with the Klein paradox (or, more generally, with the transitions to states of negative energy that is also required by the theory up to now, which we shall not go into further here) cannot be regarded as satisfactory. Initially, the mere allusion to the fact that, according to (36), D will be unimaginably small for all electric fields that can be produced in practice does not seem sufficient to invalidate the difficulty. Namely, if an electron with negative kinetic energy were present at some point then from the theory it should be just as stable as an ordinary electron, and since arbitrarily long times are available in nature, if the theory is valid then those electrons with negative masses must certainly be present in noticeable numbers, even when their probability of existence is very small.

Therefore, up to now, two attempts have been made to alter the theory in such a way that the transition to states of negative energy would be not only unlikely, but impossible. However, difficulties that are just as great appeared in other places for both attempts of that kind. Therefore, e.g., SCHRÖDINGER's proposal ([2]) (if one ignores the fact that relativistic invariance of the theory is lost in it, as SCHRÖDINGER himself emphasized) contradicts the classical Thomson formula for the scattering of light by free electrons, whose validity is required unconditionally for wave lengths of light that are large in comparison to $h\,/\,mc$ on empirical, as well as theoretical, grounds. On the other hand, in

([1]) F. SAUTER, Zeit. Phys. **69** (1931), 742; **73** (1931), 547.
([2]) E. SCHRÖDINGER, Berl. Ber. (1931), pp. 63.

DIRAC's so-called "hole world" ([1]), the second of the aforementioned attempts at an alteration of the theory, in its most recent form, an "anti-electron" is introduced in place of the electron with negative mass whose charge is $+e$ and whose mass is equal to that of the ordinary electron. It is essential for that theory that the laws of nature should be exactly symmetric with respect to the electron and the anti-electron. Therefore, in order to explain the actual absence of an anti-electron, one must make special assumptions about the initial state in nature, in which the one kind of electron must already predominate over the other one very strongly. Even if one overlooks the radiation-theoretic difficulties in that theory completely, that still seems to be an exceedingly artificial and unsatisfying solution.

Certainly, the problem of the states of negative energy is connected so closely with the question of the stability of the electron and the relationship between electrons and protons that the alleviation of that difficulty can be first expected in a theory that exhibits a logical link between the atomistic conception of electric charge and the existence of the quantum of action ([2]). On the other hand, the states of negative energy seem to this author to be so closely linked with the entire formalism of the present theory (which deals with continuous wave functions in space and time whose squares determine the probability for the position of the particle) that in the Klein paradox, we can glimpse a hint of the possibility that the future theory of the electron will be coupled with essential far-reaching fundamental changes to the formalism of the present theory of wave mechanics.

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([1]) P. A. M. DIRAC, Proc. Roy. Soc. London A**126** (1930), 360; **133** (1931), 60.

([2]) The necessity of such a link was emphasized by BOHR quite vehemently (cf., e.g., his FARADAY lecture, J. Chem. Soc., Feb 1932, pp. 349).